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PALOMARES EXPERIENCE IN PU ENVIRONMENTAL BEHAVIOUR

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Abstract

The objective of this paper is to present the most important activities carried out in Palomares in order to characterise the plutonium contaminated site, and to gain further knowledge of the plutonium behaviour in the area.

The behaviour of plutonium and americium associated with the soil for more than 30 years has been studied. These studies included chemical and mineralogical associations of plutonium with the different fractions of the soil, lung solubility of plutonium associated with the breathable fraction and statistical calculation of the inventory of plutonium remaining in the first 45 cm of the most contaminated area, in which drumping work was done. Other studies focused on the determination of the isotopic relations of plutonium and americium in the surface level are being performed.

Results show a large variation in plutonium particle sizes in the soil from Palomares. Theoretical calculations estimate that plutonium remaining in the area could be 16 per cent of the total content in two bombs. Secuential extraction techniques show that the greatest mobility stage of plutonium represent 0.2 per cent of the total extracted, being the 93 per cent content in the residual fraction. The particles show very low solubility in simulated lung fluid, and the enhancement factor, defined as the ratio of activity concentration in the inhalable fraction to that in the bulk parent soil was found to be 0.26

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1. INTRODUCTION: PURPOSE AND SCOPE

The purpose of this paper is to present the most important knowledge about the behaviour of the Pu contamination in the Palomares area (Almería, Spain), which occurred in 1966 as a consequence of an aerial accident of two USA army planes during a refuelling operation.

Two thermonuclear bombs, of the four which were transported by a B-52, had problems with their parachute, and part of their nuclear fuel was disseminated. Consequently, 226 Ha of urban, agricultural and uncultivated land were contaminated with different activity levels.

Immediately after the accident and before the remedial actions were taken alpha surface contamination levels were measured with proportional counters PAC-15 alpha detectors. A called "zero" line was defined and some study plots were established from which soil and vegetation samples were taken periodically for radioactivity analysis.

The decontamination and remediation works of the zone consisted of removing part of the soil as well as of high contaminated vegetation [1]. Once these tasks were finished, the wastes produced were sent in drums to the USA. The temporal storage and wrapping of the drums were carried out in one of the contaminated areas, which according to its natural characteristics (hills with scarce crops) was considered as the most suitable; this zone, therefore, remained with higher contamination levels than the rest.

A Environmental and personal Surveillance Program was established after the remediation works were finished. [2].

Several human activities since the accident have significantly altered the landscape at Palomares. The intent of the changes was to increase the total area available for cultivation, to store more water for crop irrigation and to conserve moisture by reducing evaporation. These activities have increased resuspension and redistribution of plutonium and americium [2].

Green houses were constructed to allow crop production during the entire year. Although these plastic enclosures have no floors, they prevent any removal of residual Pu contamination from the soil by wind action and resuspension. However, because the structures retain moisture and have no floors there exists the possibility of a change in the Pu soil chemistry with increasing time.

Between April and October 1986 a cooperative reservoir was constructed to hold 151,600 m^3 of water for irrigation. Another pond was constructed between February and May in 1988 bringing the total storage capacity to about 347,000 m^3. These reservoirs will now provide the water for the farmers, if allowed, to extend farming down to and beyond impact point 2. Also,

the topography of the land in area 2 was significantly altered as the land owners redistributed soil removed from hillsides to increase the amount of level land available for cultivation.

Since 1986, the behaviour of Pu and Am associated with the soil for 20 years has been studied. [3]. The first goal of our group is to study and to characterise the contamination, including chemical and mineralogical associations of Pu with the different fractions of the soil. Another goal is to investigate lung solubility of Pu associated with the breathable fractions.

One new statistical calculation of the inventory of Pu remaining in the first 45 cm of the soil in area 2 was made using data of Pu radiochemical analyses. Other studies dealing with the determination of the isotopic relations of Pu in the surface level are being performed.

All these works have been carried out by scientist and technicians of the Terrestrial Transuranides Environment Impact Laboratory (CIEMAT), with the collaboration of Drs. Pedro Rivas and Berta de la Cruz from the Hydrogeochemical Site Characterisation Department (CIEMAT), and the valuable help and support of Dr. Ch. Richmon from DOE (USA).

2. SOIL CHARACTERIZATION

2.1. Soil Particle Size and Activity Measurements

Distribution of particle sizes in Palomares soils as studied by several techniques show a large variation. Iranzo et al. [3] showed gross radioautographic evidence of the large variation in Pu particle sizes in soil from Palomares.

Soil samples taken in 1981 to a depth of 15 cm from several study plots were measured for ^{241}Am and Pu. Each sample was treated by maceration and ultrasonic separation and then divided into eight size fractions; 0-5, 5-10, 10-20, 20-40, 40-63, 63-250, 250-1000 and >1,000 μm. Fifteen gram samples were measured for ^{241}Am using low-energy photon spectrometry with intrinsic germanium detectors and Pu using alpha spectrometry. [3]

The $^{239+240}$Pu /^{241}Am ratio was almost constant with a large coefficient of correlation within each size fraction except for the 5-3B plot sample. The activity mass concentration for both Am and Pu was highest in the 63-250 μm fraction except for plot 5-3B where the maximum activity mass concentration occurred in the 0-5 μm fraction [2].

An association between the mineral components of the soil and concentrations of Pu and ^{241}Am was found using autoradiographic analyses. The main concentrations of alpha tracts were found in fragments of iron oxy-hydroxide and, to a lesser extent, in carbonate (dolomite) and muscovite-illite fragments. The potential leaching of Pu estimated from concentrations measured in the maceration-elutriation water was in the range of $2 \cdot 10^{-4}$ to $21 \cdot 10^{-4}$ percent of the total soil Pu. The larger values were associated with the most oxidized and least alkaline soils. Of particular interest, soil in plot 5-3B, which was not contaminated by the original accident, showed 57% of the sample activity in the < 5 μm fraction.

The particle size distribution of soil collected in November 1986 close to impact point 2 was reported for various soil fractions [4]. The samples were subjected to maceration and ultrasonic dispersion. From each sample, eight size fractions were obtained in the following size intervals, 0-5, 5-20, 20-40, 40-63, 63-125, 125-250, 250-1,000, and >1,000 μm. The silt-size particles (5 to 63 μm diameter), and the sand-size particles (63 μm and larger) accounted for 55% and 36%, respectively, of all the particles; the remaining 9% of the particles were less than 5 μm diameter. Twenty percent of the soil particles were 10 μm or less in diameter; these 5 and 10 μm diameter size ranges are generally associated with the fraction retained in the alveolar lung after inhalation. The actual deposition value is density dependent being smaller for higher density particles.

The Pu contents of the eight particle size fractions were reported for these soil samples [4]. Of the total Pu activity the silt-size particles (5 to 63 μm diameter), and the sand-size particles (63 μm and larger) accounted for 63% and 35% respectively, of all the Pu; the remaining 2% of the activity was in the <5 μm diameter particle. The average Pu concentration in the total sample was 1,944 $Bq \cdot g^{-1}$. The concentration values range from 254 to 6,389 $Bq \cdot g^{-1}$ (Table I). The observed bimodal distribution showed peaks at 3,370 $Bq \cdot g^{-1}$ for particles between 20-40 μm and 6,389 $Bq \cdot g^{-1}$ for the 125-250 μm particles. These size ranges represent 42 and 15% of the total Pu in the soil sample.

The 125-250 μm fraction is of particular interest as it accounts for 15.39% of the total Pu yet only 4.60% of the mass of the soil sample. Because of the high Pu concentration (6,389 $Bq \cdot g^{-1}$) in this fraction, the Pu concentration was studied as a function of soil density and magnetism for components of this fraction. Three densities were chosen; <2.89, 2.89 to 3.3 and >3.3 $g \cdot cm^{-3}$. For the lowest and intermediate densities, six magnetic fractions were chosen and four were chosen for the highest density fraction. Pu concentration increased with increasing density; 1,711, 4,707 and 116,379 $Bq \cdot g^{-1}$ for the lowest, intermediate and highest densities, respectively. The total Pu was divided as follows; 29, 5, and 65% for the lowest, intermediate and highest densities, respectively. (Table II) In general the Pu concentration increased with magnetic class in the lowest and intermediate densities. In the highest density fraction, the maximum Pu concentration of 502,471 $Bq \cdot g^{-1}$ was found for the lowest magnetic susceptibility.

The chemical and mineralogical composition of the soil have also been reported. The soil has a siliceous-carbonic nature; its main components, comprising 87% of the total, are SiO_2, Al_2O_3, CaO and CO_3^{2-}. There is a general tendency for quartz and feldspars to decrease and carbonates and phyllosilicates to increase as particle size decreases. As shown in Table III, dominant minerals in the gross soil sample are quartz (39%), carbonates (37%) and plagioclase (13%); the carbonates include calcite (30%), dolomite (7%) and a small amount of gypsum (<0.5%). The following account for 94% of the minerals in the < 5 μm diameter particles: calcite (58%), plagioclase (11%), quartz (10%), mica (8%) and chlorite (7%) [4]. The 63-125 μm particles are high in quartz (47.39%) and calcite (27.42%).

2.2. Geochemical associations of plutonium in soil

In order to gain further knowledge of the geochemical associations that Pu show in the soil, a sequential leaching method has been applied to a contaminated soil from Palomares. The method employed is basically McLaren and Crawford, and modified by Cook et al. [4], applying modifications brought in by our work group[5], see table IV.

The procedure was carried out in seven consecutive stages using two samples of the same soil, each weighting five grams. The soil activity for $^{239+240}$Pu is 1727 $\pm$ 180 Bq·gr^{-1}. ^{239}Pu/^{238} Pu ratio is around 54 (1992).

The results obtained on $^{239+240}$Pu in the different soil fractions are shown in table V. The greatest mobility stage of $^{239+240}$Pu, made up of the soluble, exchangeable and inorganically adsorbed fractions, represents 0.2 per cent of the total ^{239} Pu+ ^{240}Pu extracted. Associated to organic matter we have 6 per cent of the total $^{239+240}$Pu , and approximately the remaining 93 per cent is associated to sesquioxides and distributed between the two residual fractions. The ^{238}Pu distribution does not vary substantially from that of the ^{239}Pu, except in the soluble and exchangeable fractions where the ^{238}Pu percentage is around three times the ^{239} Pu percentage, which brings about a variation in the ^{239} Pu/^{238}Pu ratio.

2.3. Lung solubility

The purpose of the experiments performed is to study the pulmonary biokinetics (by means of simulation) of plutonium and americium associated with contaminated soil. The methodology used takes into account different fractions of soil: $\Phi < 5$ μm (respirable fraction), 20-40 μm, 125-250 μm and total soil. For pulmonary simulation small amounts of each dust sample were placed in a tube-shaped, semipermeable membrane closed at both ends. Each membrane was placed in a plastic bottle with 400 ml of pH 7.3 physiological solution. Every bottle was kept at 37°C and was shaken for half an hour every three hours so as to simulate the physiological conditions of the lung. Each week the bottle was replaced by another with fresh physiological solution, and the solution was analysed using radiochemical techniques.

The chemical composition of physiological solution used contained mineral salts (ClNa, NH_4Cl, NaH_2PO_4, ClCa), amino acids (Glicine, Na_3Cit) and H_2SO_4.

The first stage of the experiment was maintained for 3 months, and during this time it was observed that the amount leached did not exceeded 0.5% of the total plutonium contained in the sample. Another simulation experiment was performed with two other alicuots of the fraction below 5 microns. During the first weeks of this experiment similar percentages of ^{239} Pu+ ^{240}Pu and ^{241}Am were disolved, but the percentage of ^{238}Pu was several times. The total amount of $^{239+240}$ Pu and ^{241}Am extracted during the first 140 days were less than 0.58 and 0.32 of the total $^{239+240}$ Pu and ^{241}Am contained in the respirable fraction of soil. For ^{238}Pu the values were about 2.

Six months after the start of the experiment, the physiological solution used in one sample was replaced by specific reagents for sequential leaching, in order to establish the

geochemical associations of the radionuclides not extracted in the simulated pulmonary leaching experiment. The method employed is basically that of McLaren and Crawford. The reagents and fractions extracted are shown in table VI.

The sequential leaching in two samples (6 months and 1 year after pulmonary experiment) with acetic acid and sodium pyrophosphates showed that the specifically-adsorbed and organically bound fraction had been reduced or even disappeared in the case of one year after the pulmonary leaching experiment. This result shows that the radionuclides in exchangeable, specifically-adsorbed and organically bound forms are the most readily leached by physiological solution. These fractions represent less than 4% of the total activity for $^{239+240}$Pu and 8% for ^{238}Pu in the total soil sample.

It is important to underline that in the pulmonary simulation experiments with the respirable fraction sample (employing 3 alicuots), the ^{238}Pu extraction, especially during the first four weeks, was higher that the $^{239+240}$ Pu extraction obtained.This phenomenon did not take place in simulation experiments with the other soil fractions. The water solubility of ^{238}Pu, when incorporated in relatively large plutonium oxide particles (>1 μm), would be expected to be greater than the solubility of $^{239+240}$ Pu oxide particles of similar size as a result of crystal damage and radiolysis arising from the greater specific activity of the ^{238}Pu (an approximate factor of 270).

The activity percentage leached in the respirable fraction experiment, during the first six weeks, is similar for $^{239+240}$ Pu and ^{241}Am. The ^{238}Pu activity percentage leached about 4 times higher than that of the $^{239+240}$ Pu, this difference being greater during the three first weeks.

The results obtained in the pulmonary leaching experiments performed with 20-40μm, 125-250 μm granulometric soil fractions and total soil show, in all cases, the similar behaviour of the plutonium isotopes. However in the experiment performed with the respirable fraction, the most soluble form was ^{238}Pu . The different behaviour of the breathable fraction of the Palomares soil, in the simulation experiments, can be explained by its different mineralogical composition (see table VII). Correlation factors between the plutonium activity concentration in each granulometric fraction and the abundance of the minerals present in each fraction were calculated for some samples of the Palomares area and are shown in table VIII. These factors show the correlation that exist between the plutonium activity concentration and the abundance of minerals like chlorite and illite-moscovite. Both of them are phyllosilicates formed by sheets, respectively 14 and 10 Amstrongs wide, fixed one to another by cations like K, Na, Fe, Mg. Plutonium and americium can principally be absorbed onto the surface of these minerals, some amount of these radionuclides could be occluded or trapped into the structure of the cristals according to the extraction with HF-HNO3 mixture realized during the sequential extraction procedure.

3. INVENTORY OF PU

3.1 Inventory of Pu estimates for zone 2-0

Resurveys of Pu contamination in some areas around Palomares have been started with the objective of provide estimates of the residual plutonium and americium in the area.

37 points were selected for taking soil samples to a depth of 45 cm in the most contaminated zone. For each sample, fractions were taken at five depths (0-5, 5-15, 15-25, 25-35 and 35-45 cm) and the resulting material was analyzed by alpha spectrometry for Pu and by low-energy photon spectroscopy for ^{241}Am. The concentration of Pu and ^{241}Am were not distributed in a homogeneous manner. Both radionuclides were found down to 45 cm depths in the valley. On the hillsides the concentrations of Pu and ^{241}Am below the first 5 cm was one or more orders of magnitude less than that in the valley. An average Pu concentration of about 82 kBq·kg^{-1} was found with a wide variation from minimum to maximum. The average ^{241}Am concentration was about 13.4 kBq·kg^{-1} with a large range of values around the mean. For both Pu and Am the highest concentrations were found near the original impact point 2. A gross estimate of the plutonium content of the 6.5 Ha was $6 \cdot 10^{11}$ Bq (about 17 Ci) [6].

A recent study of a select 5.25 Ha (52,500 m^2) site in area 2 suggests an inventory of $2.86 \cdot 10^{12}$ Bq (77 Ci Pu) to a depth of 45 cm [7]. The data are from surveys conducted in 1986. At 37 locations Pu samples were taken at six depths down to 45 cm fig. 1. Various statistical procedures were employed to calculate the volume, mass and Pu concentration of each soil layer (0-5, 5-15, 15-25, 25-35 and 35-45 cm), therefore the entire 5.25 Ha down to 45 cm. The relative soil density was found to be 1.6 g·cm^{-3}. The $2.86 \cdot 10^{12}$ Bq (77 Ci) inventory represents a mass of about 1.3 kg Pu. Although the area covered is somewhat limited, it is thought to represent the most contaminated land contained within area 2. Calculations indicate that removal of the top 5 cm of soil, containing $1.45 \cdot 10^{12}$ Bq Pu and weighing about $4.1 \cdot 10^{3}$ metric tons, would remove 51% of the total inventory. Figure 1 shows the location of the study area and the residual Pu concentrations in the soil.

This $2.86 \cdot 10^{12}$ Bq inventory represents about 33% of the Pu in a nuclear weapon assumed to contain 4 kg Pu. However, we have to take in mind that Pu contaminated soil from the weapons in both areas 2 and 3 was brought to area 2, within the 5.25 Ha studied, until disposed of in 200 L steel drums.

Estimates of total Pu released from weapons 2 & 3 are very uncertain. The amount of Pu originally in weapons 2 and 3 is classified and we do not know how much Pu was removed from Spain for disposal in the U.S. Neither do we know with a high level of confidence the amount of Pu remaining after the initial remediation in 1966. We do know that, based on the relatively crude measurements made by the PAC 15 detectors, Pu concentrations below $1.2 \cdot 10^{3}$ Bq·m^{-2} were plowed into the soil to reduce the original surface concentrations.

3.1. Plutonium vs. Americium

Taking into account that the isotopic composition of Pu in the Pu weapon grade has an activity ratio ^{241}Pu vs $^{239+240}$Pu at least of 5 and that ^{241}Am comes from the decay of ^{241}Pu, the activity concentration of Am is expected to increase with time. In order to estimate the correlation between Pu and Am, 45 samples from the studied area have been used, all of them with activity values of both radionuclides much higher than the LID. These data are plotted in fig. 2, in which Pu as well as Am are not homogeneously distributed, though the highest concentration in both cases are situated in the same points.

The linear correlation of $^{239+240}Pu$ vs. ^{241}Am is shown in fig. 3, the correlation coefficient being greater than 87%. Future works will be focused on the obtention of isotopic relations of Pu and Pu/Am in different types of samples to be able to determine possible similarities or differences in the behaviour of these radionuclides.

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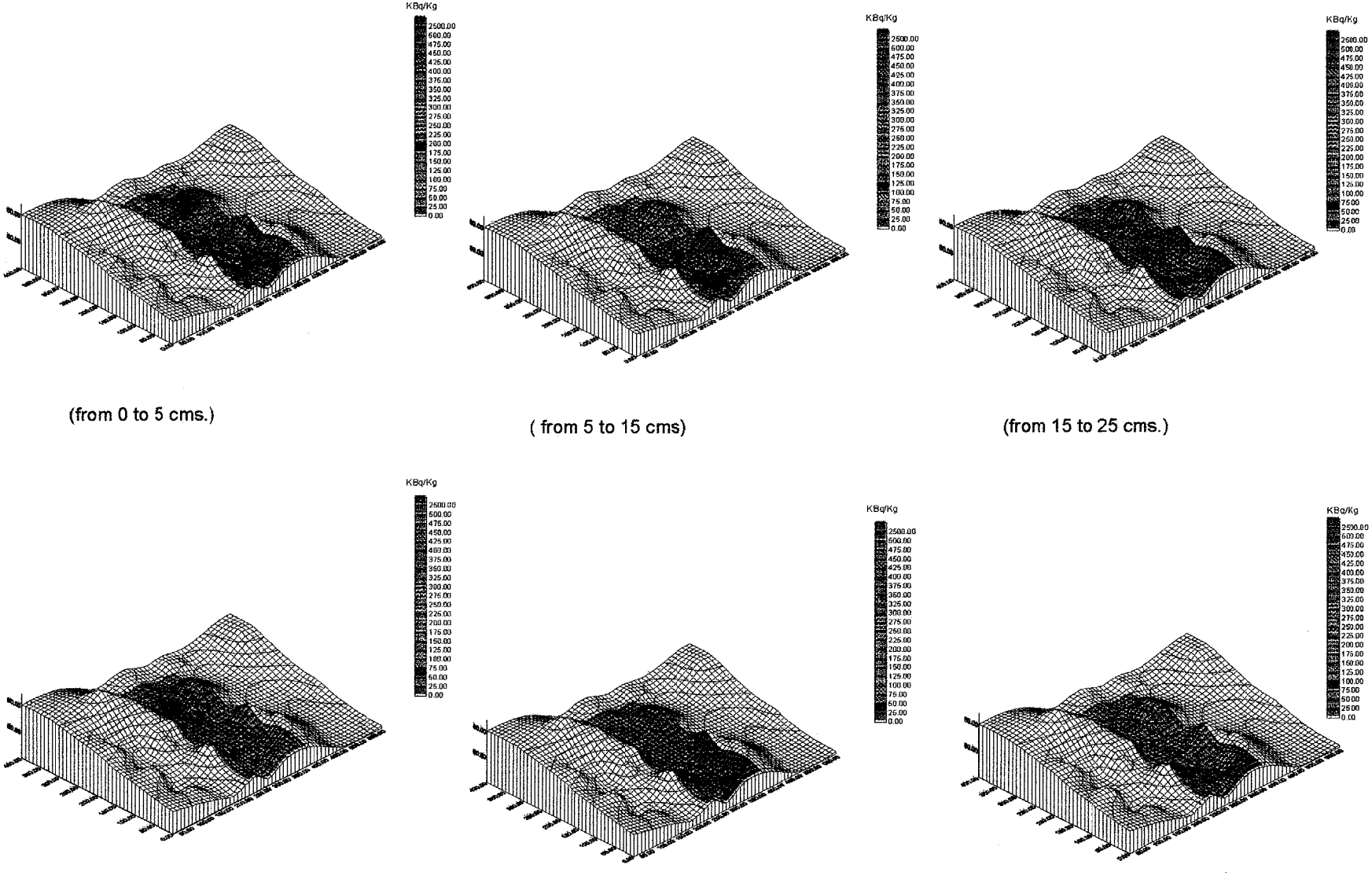

Fig.1 The location of the study area and the residual Pu concentration in the soils

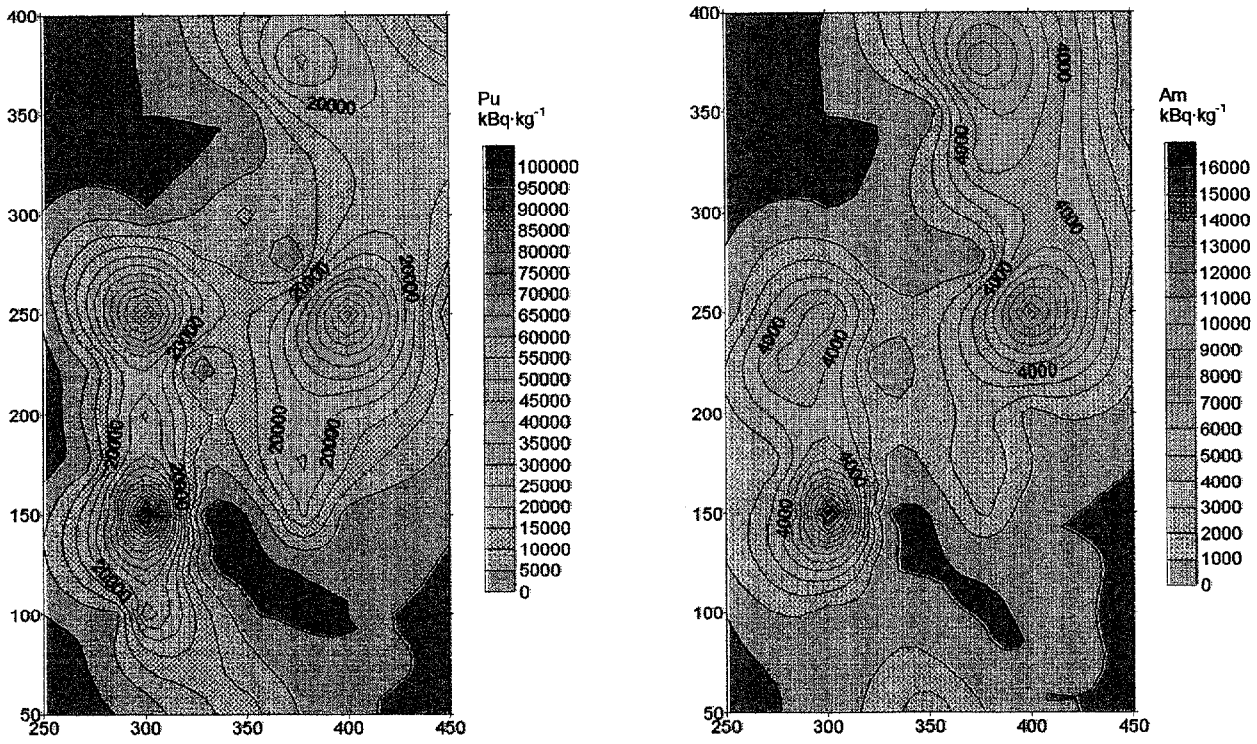

Fig. 2. Contour maps of plutonium and americium

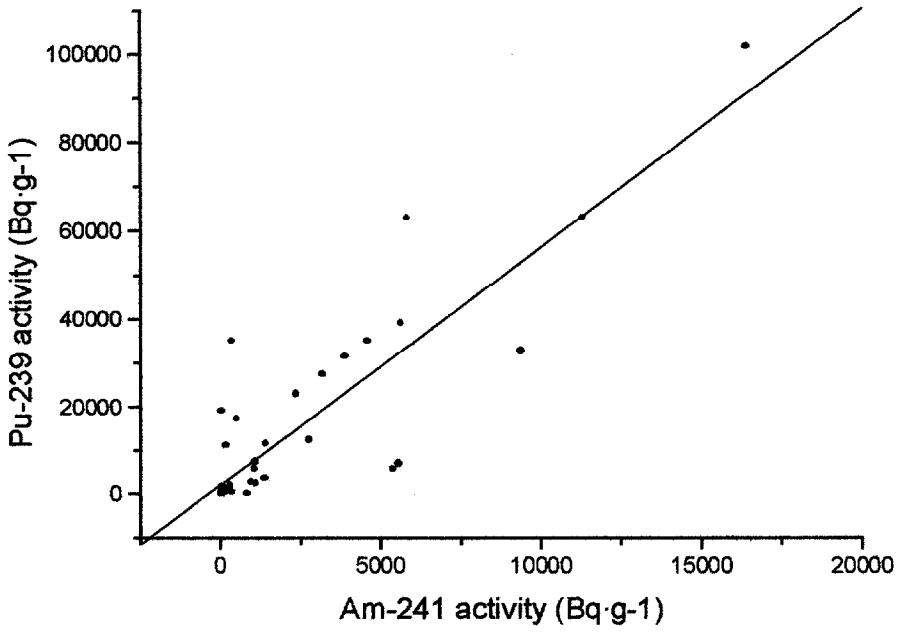

Fig. 3. Linear correlation. Pu activity vs. Am activity in soil samples

TABLE I. PLUTONIUM ACTIVITY IN THE PARTICLE SIZE FRACTIONS OF SOIL

SIZE CLASS μm	ABUNDANCE (%)	Pu $Bq \cdot g^{-1}$	Pu Abundance (%)
5	8.78	504±14	2.32
Silts			
5-10	11.40	1333±79	7.96
10-20	8.77	1056±67	4.85
20-40	23.73	3370±210	41.37
40-63	10.77	1504±82	8.48
Sands			
63-125	13.79	2183±99	15.76
125-250	4.60	6389±315	15.39
250-1,000	2.87	904±42	1.36
>1,000	15.19	2544±9	2.02

TABLE II. PLUTONIUM ACTIVITY DISTRIBUTION AS A FUNCTION OF DENSITY FOR 125-250 MICRON DIAMETER SOIL PARTICLES.

DENSITY $G \cdot cm^{-3}$	**WEIGHT ABUNDANCE** %	**Pu** $Bq \cdot g^{-1}$	**Pu** %
<2.89	91.0	1,711	29.3
2.89-3.3	6.0	4,707	5.3
>3.3	3.0	116,379	65.4

TABLE III. MINEROLOGICAL COMPOSITION OF THE TOTAL SAMPLE AND SEVERAL SELECT SIZE FRACTIONS

MINERAL	% OF SAMPLE	125-259 μm	63-125 μm	5-10 μm	5 μm
Quartz	38.61	59.75	47.39	12.45	10.28
Carbonates	37.55[a]	9.03	29.71	53.09	62.22
Plagioclase	13.46	15.69	12.29	15.66	10.74
Others[b]	10.38	15.53	10.61	18.80	16.76

[a] Calcite (30.09%), Dolomite (6.97%) and Gypsum (0.49%)
[b] Mica, Chlorite and Orthoclase

TABLE IV. APPLIED SECUENTIAL LEACHING PROCEDURE.

Fraction obtained	**Reagent**	**Volume/soil ($ml \cdot g^{-1}$)**
Water soluble	Distilled water	20
Exchangeable	0.005M CaCl2	20
Specifically adsorbed	0.05M Acetic acid	20
Organically bound	0.01M Na4P2O7	100
Oxide bound	0.175M (C02NH4)2/ 0.1M(C02H2)2	75
Residual	7.8M HN03	70
Residual (strongly bound)	HF + HN03	70

TABLE V DISTRIBUTION OF PLUTONIUM IN THE VARIOUS FRACTIONS OF THE SEGUENTIAL LEACHING PROCEDURE

Fraction obtained	**Pu239+Pu240(%)**	**Pu238(%)**	**Pu239/Pu238**
Water soluble	0.0036	0.01	19
Exchangeable	0.0054	0.013	22
Specifically adsorbed	0.2	0.18	55
Organically bound	3.6	7.8	50
Oxide bound	34.3	33.9	55
Residual	31.7	30	58
Residual (strongly bound)	27.8	28.1	54

TABLE VI. SECUENTIAL LEACHING PROCEDURE FOR LUNG SOLUBILITY SIMULATION

FRACTION EXTRACTED	REAGENT
Specifically adsorbed	0.05M Acetic acid
Organically bound	0.01M $Na_4 P_2 O_7$
Oxide bound	0.175M $(CO_2NH_4)2$/0.1M $(CO_2H_2)_2$
Residual	7.8M HNO_3
Residual (strongly bound)	HF + HNO_3

TABLE VII. MINERALOGICAL COMPOSITION OF SIZE FRACTIONS

Size μm	Chlorite d(Å)-14	Illite Moscovite d(Å)-10	Quartz d(Å)-4.24	Calcite d(Å)-3.03	Dolomite d(Å)-2.88
< 5 μm	67.39	46.48	6.65	100.00	5.42
5 < Φ > 10	58.70	72.24	11.97	76.78	10.54
10< Φ > 20	58.70	59.62	21.51	61.30	7.53
20< Φ > 40	84.78	97.18	55.43	65.33	24.90
40 < Φ > 63	62.22	100.00	58.09	39.00	100.00
63 < Φ >250	80.43	93.43	84.92	49.54	54.81
250< Φ>1000	41.30	33.80	100.00	73.99	19.58

TABLE VIII. MULTIPLICATIVE COEFFICIENTS OF CORRELATION BETWEEN THE ACTIVITY CONCENTRATION OF Pu-239+240 AND THE MINERALS.

SAMPLE	CLORITE	MOSCOVITE	QUARZ	CALCITE	DOLOMITE
1	0,82	0.90	-0.03	-0.56	0.45
2	0.85	0.90	0.31	-0.49	0.50
3	0.87	0.88	0.03	-0.55	0.50
4	0.97	0.74	-0.08	-0.15	0.21
5	0.75	0.41	-0.13	0.03	-0.07